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13. ABSTRACT *(Maximum 200 words)*

We find that spectral hole burning in a 2-D layer of connected Microparticles (size < 2 μm) by a plane wave is the sum of an intrinsic effect due to Morphologically Dependent Resonances (MDRs) and an artifact associated with holographic patches produced by the interference of scattered radiation with the incident wave. The effects of interparticle dielectric interaction and scattering are even more pronounced in simple 3-D clusters. <u>A new excitation configuration has been invented which involves the coupling of microparticles to a thinly clad fiber</u>. In this configuration MDRs are selectively stimulated on the basis of the correspondence between their angular momentum and impact parameter associated with the distance of the fiber axis fromthe center of the sphere through the "Principle of Localization." Simple models have been devised to describe this coupling with and without the fiber coupler index matched to the medium surrounding the sphere. <u>The fiber - MDR coupling (FMC) mechanism is expected to be a harbinger which will allow the introductionof compact high Q sperical resonators into active and passive photonic devices.</u>

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ROOM TEMPERATURE MICROPARTICLE BASED PERSISTENT SPECTRAL HOLE BURNING

STARTING DATE: 15March 1994

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Principal Investigatror

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I. INTRODUCTION and BACKGROUND

Although Room Temperature Persistent Spectral Hole Burning (RTPSHB) has been a dream of many scientists working in information storage and material science for the last 21 years, it has only had isolated and modest success in the last four years.[1] Efforts to create amorphous materials (principally glasses[2]) in which to burn holes at high temperatures face an inherent contradiction: the need for inhomogeneous line broadening from host-guest interactions, vs. the desire to limit the homogeneous line broadening from thermal fluctuations of host-guest interactions (i.e. phonon broadening). In light of this contradiction we have taken a new approach.

We have demonstrated RTPSHB using a 2-D collection of fluorescent spherical microparticles having a random distribution of sizes.[3] In this system, known as a Microparticle Hole Burning Medium(MHBM), the differences in the frequencies of Morphology Dependent Resonances(MDR)[4] of individual particles with size enables one to generate a fluorescence excitation spectrum which is heterogeneous.

MDRs are associated with photon confinement by a particle's "dielectric potential". In fact one can transform the usual electromagnetic vector wave equation into a Schrodinger equation which reveals modal functions which are analogous to wavefunctions of electrons in a Rydberg atom; the photon circumnavigates the particle, near the surface. However, unlike states in a conventional atom the modes of a "photonic atom"[5] are virtual with the photon lifetime limited by leakage out of the particle.[6] The leakage can be extremely slow. Recent measurements reveal Q's approaching 10^7 in particles only ~ 5 μm in radius.[7,8] Furthermore the theoretical analysis shows that each resonance occurs at a constant value of ka, where k is the magnitude of the wavevector in vacuum and a is the particle radius. Thus a collection of particles having a distribution of sizes of width σ_a when irradiated near a given resonance should display an inhomogeneous width $\gamma_{ih} \approx \sigma_a \langle k \rangle / \langle a \rangle$, where $\langle k \rangle$ is the average wavevector of the radiation and $\langle a \rangle$ is the average particle size. From the stand point of hole burning the advantages of using a collection of particles as a hole burning memory are

a. the homogeneous linewidth associated with leakage $\gamma_h \approx \langle k \rangle / Q$ is narrow and virtually insensitive to temperature, and
b. the inhomogeneous linewidth $\gamma_{ih} \approx \sigma_a \langle k \rangle / \langle a \rangle$ associated with the size distribution width can be much broader than γ_h since it is controlled independently.

In what follows we outline the direction taken by our research in connection with AFOSR Grant F49620-94-0195 (Sec.I I).

II. DIRECTION OF RESEARCH

Since the particles in the original study were large in comparison to the pits in a contemporary optical storage medium (areal density of $\sim 10^6$ cm^{-2} vs 10^7 - 10^8 cm^{-2}), aside from the obvious gain associated with multiple spectral addressing, MHBM can only be competitive by using smaller particles or higher dimensional arrays. Therefore we began our current investigations, in connection with MHBM, by performing experiments on 2-D layers of smaller particles (<12 μm, Sec III. a). We found as the size of the particles was reduced below 2 μm artifact holes were produced which were not due to MDRs, but most likely resulted from "holographic" patches associated with interference between the incident plane wave and scattering within the layer. This led us in the direction of asking whether particles placed behind one another could be separately addresses. Here the effects of multiple scattering were even more pronounced as evidenced by the results of fluorescent excitation spectra on multiparticle clusters (Sec III.b). Although, a Ph.D. thesis was constructed from this research (the 1st optical spectroscopy of a microsphere cluster),[9] it became apparent that a means had to be devised for exciting MDRs while reducing multiple scattering. Fortunately, such a means was devised. By placing particles on a thinly clad optical fiber, we were able to selectively address only MDRs and avoid interparticle scattering. Most of our research focussed on investigating this approach (Sec III.c).

III. RESEARCH ACTIVITIES

A. 2-D Arrays of Particles Smaller in Radius than 12 μm.

Fig.1 shows burn spectra for a film of particles $\sim$ 5 μm in radius. The holes were burned into the film at 1W/cm^2 for 15 sec, and each spectrum was read out over 1 min at 1/100 of this intensity. The spectra were taken with the laser projected onto the film at 45 degrees to the normal, and the fluorescence was detected in a backscattering configuration. The spectra were taken 10 minutes apart. The holes are distinct and persistent. However, as the particles were reduced below $\sim$2 μm in radius the spectra developed an angular dependence indicating that at these sizes the photolysis was becoming non-local (not entirely associated with the stimulation of MDRs in the individual particles); "holographic" patches were most likely being generated by interference between the incident plane wave and scattering within the layer.

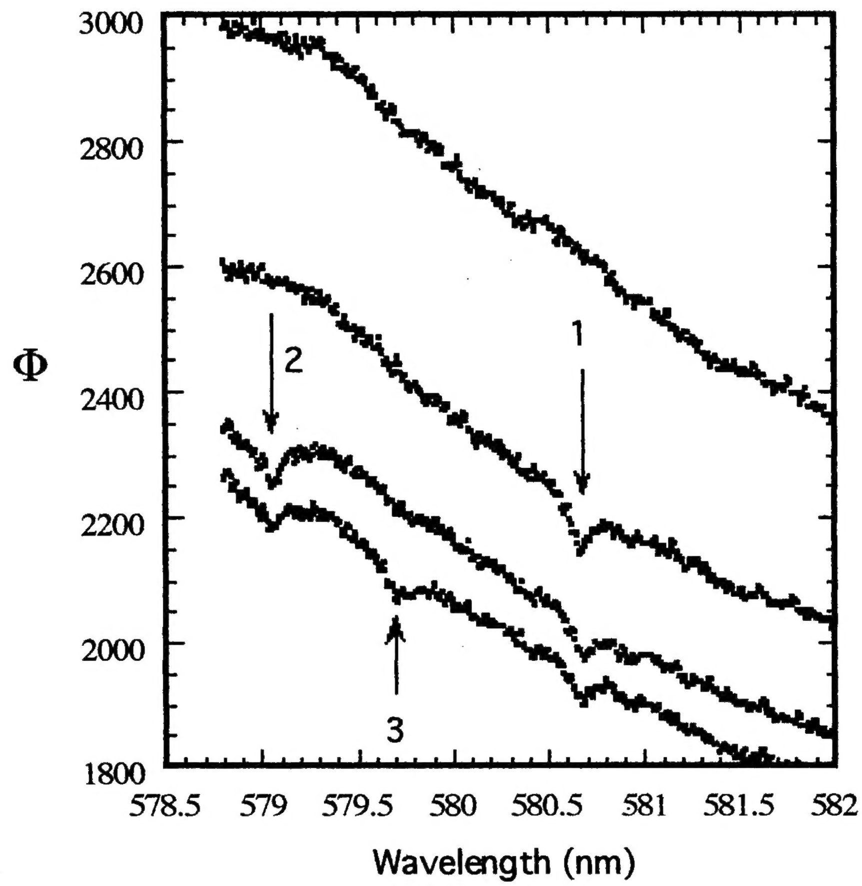

Fig. 1 Successive hole-burning fluorescence excitation spectra for a 2-D layer of polystyrene particles 5μm in radius.

It became apparent that inorder for microparticle hole-burning to be viable the particle had to be excited in a manner in which multiple scattering could be reduced in comparison to the excitation of MDRs

B. Two Particle Clusters.

Experiments were performed on isolated two particle clusters (polystyrene spheres, each of radius $a \approx 4.5\ \mu$m) in an attempt to ascertain whether particles in contact with others could be individually addressed.[9] The clusters were isolated by levitation in an electrodynamic levitator-trap as shown in Fig. 2.[10] When the cluster was irradiated perpendicular to the line of centers its excitation spectrum contained many individual resonances associated with each particle as had been predicted by theory.[5] However, when irradiated along the line of centers at least half of the resonances were missing, and those which appeared were substantially

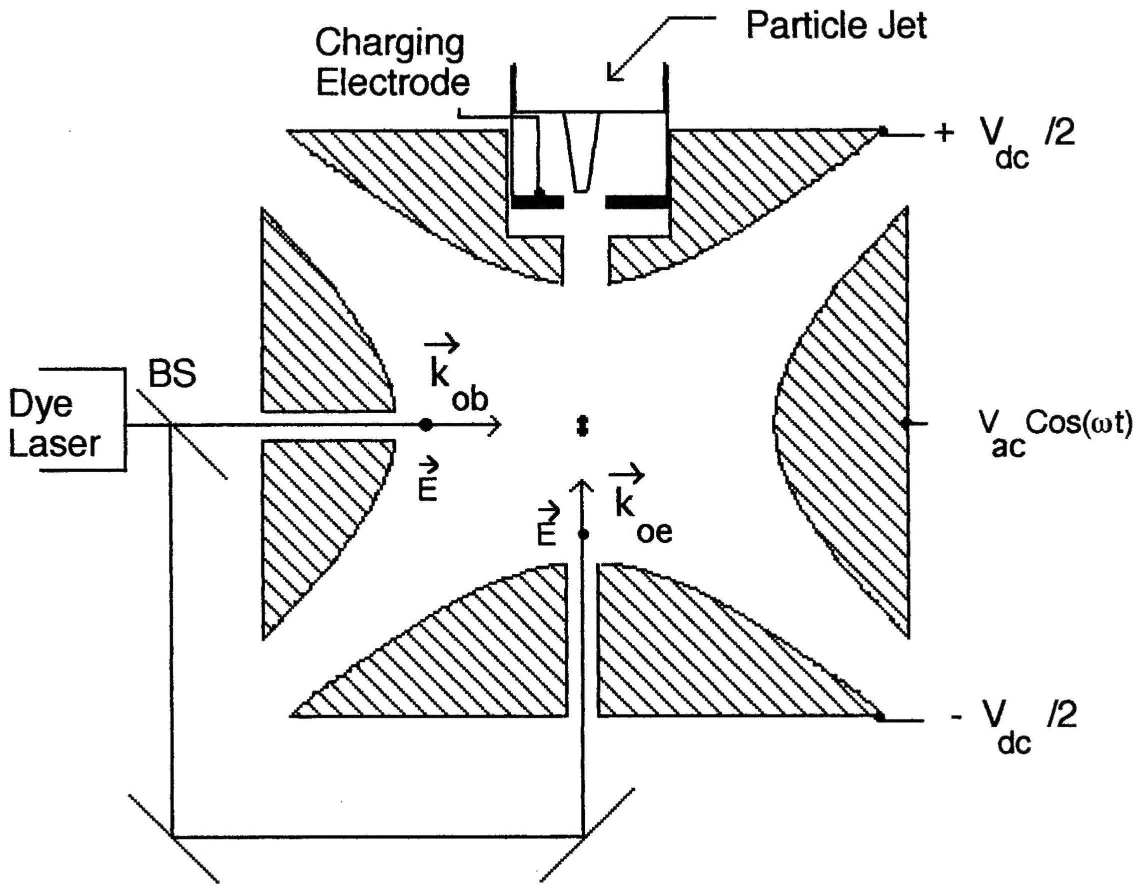

Fig.2 Experimental setup for obtaining fluorescence excitation spectra on 2 particle clusters.

broadened. These experiments indicated that layering to improve areal density would not be viable for plane wave illumination.

C. Single Particles and Multiple Particles on a Thinly Clad Optical Fiber.

To understand how to eliminate multiple scattering, one must take a careful look at the problems associated with using extended plane waves. Fig. 3 shows ray paths associated with plane wave illumination. Only a few ray paths are shown, but they are typical of rays with impact parameters b less than the radius a. A few of these paths are evidenced by viewing the particle at right angles with a microscope. When the particle is a few microns in size, its image is composed of just three "glare spots" (i.e. 1-3 in Fig. 3) .[11] Significantly, none of the incident rays couple to resonances (MDRs),

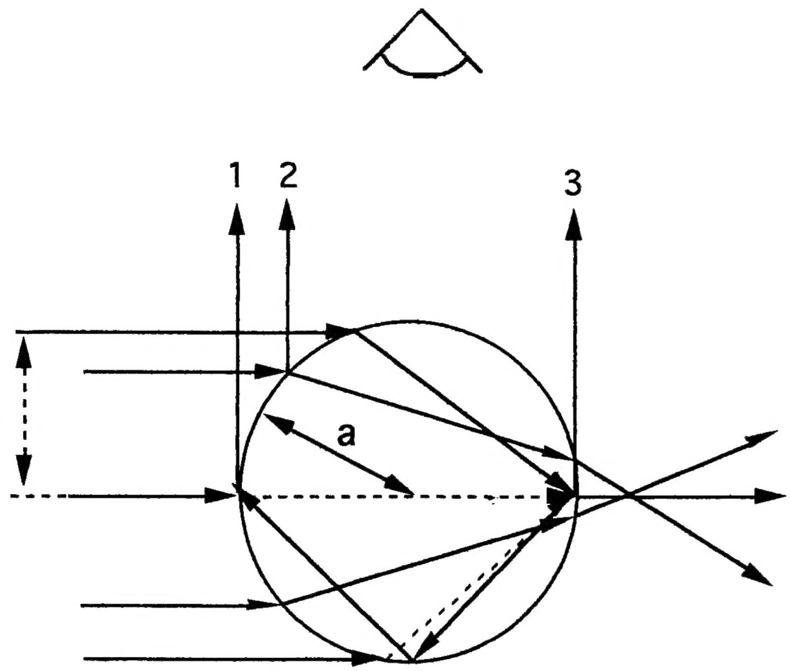

Fig. 3 Ray paths associated with plane wave illumination.

although each leads to energy transmitted through the particle, and to the photolysis of dye. In addition the emerging rays from an individual particle multiply scatter off neighboring particles to the side and directly behind, leading to further damage. In fact, no portion of the plane wave illuminating the interior of the sphere can excite an MDR. MDRs are excited by rays with impact parameters, b, which are larger than the radius a. This is most easily understood by equating the angular momentum in an MDR to the angular momentum of the external ray which stimulates the mode.

Fig. 4 shows a ray with wavevector **k** impinging on a particle in which it excites an MDR. The MDR is represented by a ray "orbit" which circumnavigates the particle while being confined by grazing "nearly total" internal reflections (i.e. there is leakage associated with rays impinging on a curved surface) . The orbit has an angular momentum $\leq \hbar k_m a$, where k_m is the propagation constant within the interior ($k_m = mk$, where m is the refractive index of the particle). Through the "principle of localization", the impact parameter for each ray has an associated angular momentum quantum number, which is equal to the orbital quantum number of the MDR (ℓ) which the ray most efficiently stimulates.[12] In effect, this allows us to equate the "orbital" angular momentum in an MDR to the angular momentum of an incident photon, $\hbar kb$. By applying this principle we find that $b \leq (k_m/k)a = m\,a$. Since m is greater than 1 for

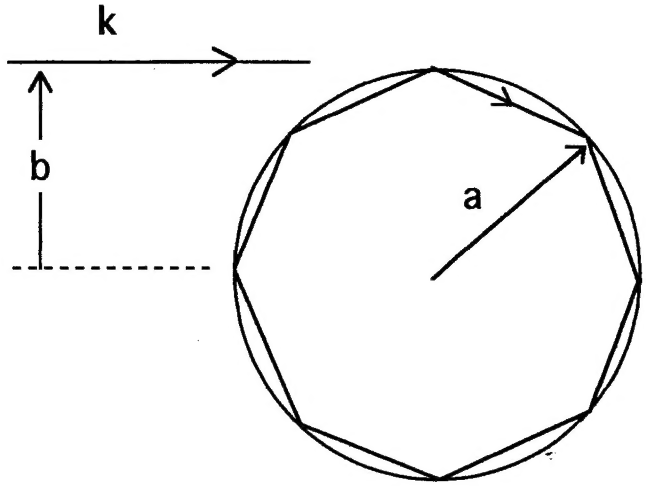

Fig. 4 Stimulation of a "geometrical" resonance by an external ray.

a dielectric particle, the incident ray will best stimulate our "geometrical" MDR for an impact parameter considerably larger than the radius! It should be pointed out that although our mode representation is geometrical (i.e. the particle size should be much larger than the wavelength), our approximate expression for the upper limit of the impact parameter b applies even when the radius is comparable to the wavelength. Other geometrical modes may be depicted by ray trajectories which close after several cycles.[12] Such a mode has a smaller angular momentum since it circulates more deeply within the interior. A complete wave analysis[13] shows that MDRs are best stimulated by impact parameters in the range

$$a < b < ma . \quad (1)$$

This inequality suggests a means for avoiding multiple scattering. Instead of using a plane wave, which leads to scattering paths which damage neighboring particles, one should use a beam to more exclusively stimulate MDR's.

The photonic analog of a beam in space is a guided wave. A particle sitting on an optical fiber might be a substitute for a beam irradiating a particle in free space (Fig. 5) if the partice's high Q MDRs could be preserved in the presence of the perturbing surface. Optically this can be accomplished by placing a barrier of lower refractive between the fiber core and the particle. Such a barrier can be effected by leaving a

small thickness of cladding in place. A description of the fiber-MDR coupling mechanism (FMC) follows.

The FMC coupling idea, as it evolved, was to shave the cladding down on a communication fiber to a thickness which would satisfy the inequality in Eqn.1. We started with particles of polystyrene nominally 12 μm in radius ($a \approx 12\mu$m). The fiber which was picked had a cladding with a refractive index of 1.475 ($m_c = 1.475$). If the particle's surroundings were matched to the index of the cladding then the refractive index m in Eqn. 1 is the relative refractive index, i.e. $m = m_{particle}/ m_c$. For polystyrene $m_{particle} = 1.59$, so $m \times a = 12.77$ μm. Consequently we decided to leave no more than ~ 0.7 μm between the cladding and the core.

The insert in Fig.5 shows the manner in which our FMC idea is put into practice.[14] A polystyrene (PS) microsphere with an approximate radius of 12 μm

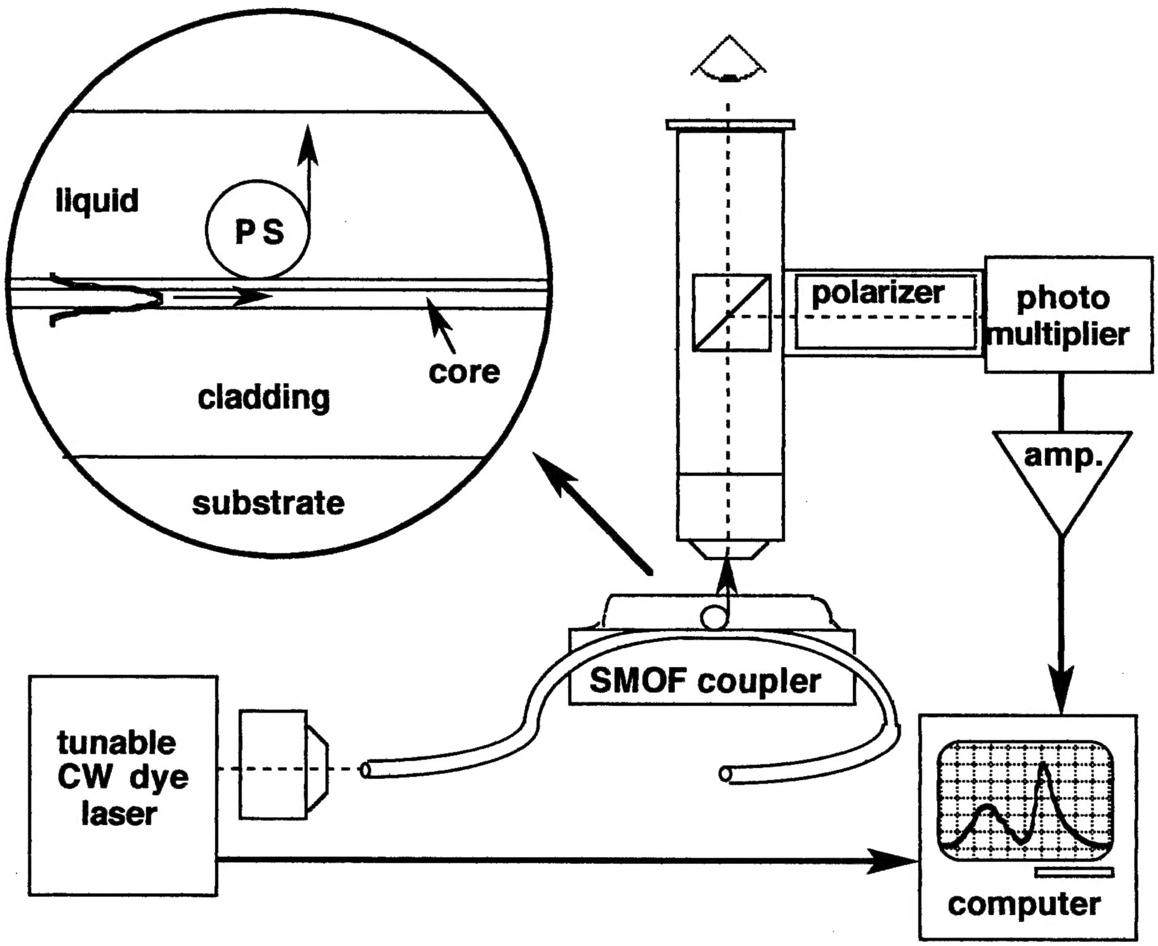

Fig.5 Setup for excitation of MDRs by a guided wave in an optical fiber.

and refractive index of 1.59 is placed on an "Optical Fiber Coupler"(OFC) using micromanipulators. Our OFC is made from a single-mode optical fiber (SMOF) with a core radius of 1.9 μm and refractive index = 1.462, and a cladding radius of 62.5 μm with refractive index m_c = 1.457. The cladding of the fiber below the microsphere is shaved down to 0.7 μm in order to maintain an optical barrier, while approximately satisfying the inequality in Eqn. 1. The SMOF mode has an approximate Gaussian intensity profile and is doubly degenerate with both linear polarization components. The OFC surface and the microsphere were wetted by a few millimeters of index matching liquid with refractive index = 1.456 (same as the cladding) to index match the cladding of the fiber, and optically eliminate the air-cladding interface at the surface of the OFC. Then the excitation geometry effectively becomes the optical equivalent of a Gaussian beam with an infinite skirt length passing near a microsphere.

The excitation light for the microsphere is provided by a tunable and linearly polarized CW dye laser with optogalvanic calibration and a linewidth of 0.025 nm.[13] The output of the dye laser is coupled to the SMOF through a microscope objective. Although the output of the dye laser is linearly polarized, the output from the SMOF is observed to be elliptically polarized due to the birefringence of the fiber. Therefore, the OFC provides both linear polarizations components for the excitation of the microsphere. The scattered light from the microsphere was collected at 90±5° through a microscope objective (with a numerical aperture of 0.17), which is followed by a polarizing prism and finally detected with a photomultiplier tube.

If a plane wave geometry were to be used for the illumination of the microsphere, we would have observed three principal glare spots through the microsphere (corresponding to rays 1-3 in Fig. 3).[11] However, in our case of coupling a Gaussian beam from the OFC, we observe only one glare spot on the far side of the microsphere.[15] In contrast to the experiments performed with non-index matching liquids,[14] this far side glare spot is observed continuously, even when the incident laser wavelength does not correspond to a MDR wavelength (i.e., off resonance). However, when the incident laser light is on resonance, this far side glare spot intensity is enhanced by a factor of two. Apparently, the standing wave pattern, with its two counterpropagating traveling waves, which is usually setup by a plane wave excitation of a MDR, is now replaced with a single traveling wave in the Gaussian beam excitation geometry. Also, in the Gaussian beam excitation geometry, the off-resonance glare spot is due to refraction, while for a plane wave illumination

geometry, the off-resonance glare spots would be due to refraction and reflection from the spherical boundary of the microsphere.

Fig. 6 shows the elastic scattering spectrum at a scattering angle of 90±5° from the microsphere obtained through a polarizer with its polarization axis at 90° to the SMOF.[15] From the polarizer orientation, we can deduce that the MDR's of Fig. 6 are of transverse electric (TE) type. The spectrum in Fig. 6 has been normalized by the laser intensity spectrum, which decreases continually with increasing wavelength. When we compare the spectrum of Fig. 6 with a scattering spectrum of a plane wave from a microsphere, we notice two noteworthy features. (1) The resonances are considerably more pronounced than in the plane wave case although there is a background which is more than the scattered light due to the OFC surface imperfections, and (2) MDR's have nearly Lorentzian lineshapes.

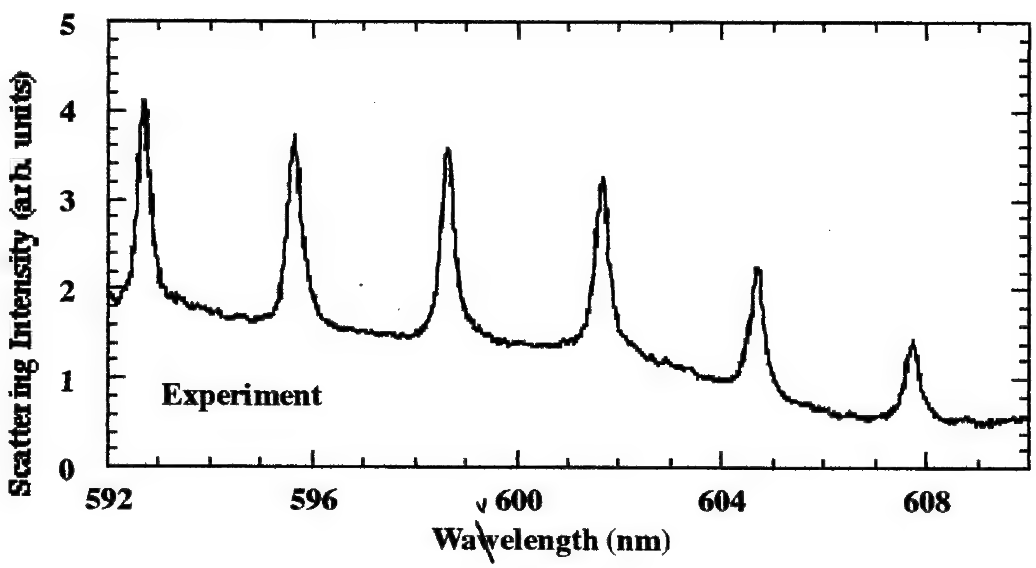

Fig. 6 TE elastic scattering spectrum taken on an index matched μ-particle 15μm in radius using fiber coupling.

True exo-beam excitation experiments of microparticles have not been previously presented although there is a theory for the effect known as Generalized Lorentz- Mie (GLMT) theory.[16] We applied GLMT to the data in Fig. 6, and were able to match resonance for resonance and also understand the background. Out GLMT calculations suggest that: (1) we should observe much narrower resonances if the dielectric contrast between the particle and its surroundings is increased, and (2) the forward scattering should be decreased by coupling to the particle. The later essentially

implies that we should observe a decrease in the intensity of light transmitted through the fiber.

To look for narrower resonances associated with enhanced dielectric mismatch we surrounded the microsphere in Fig.5 with water. This increased the dielectric mismatch (the ratio of refractive index of the particle to the refractive index of its surroundings) from 1.09 to 1.195. Fig. 7 shows the resulting spectra for two orientations of the output polarizer (Fig.7 a selects only TE modes whereas Fig. 7 b selects both TE and TM modes). Now the Q's of ~10^3 which were measured in the

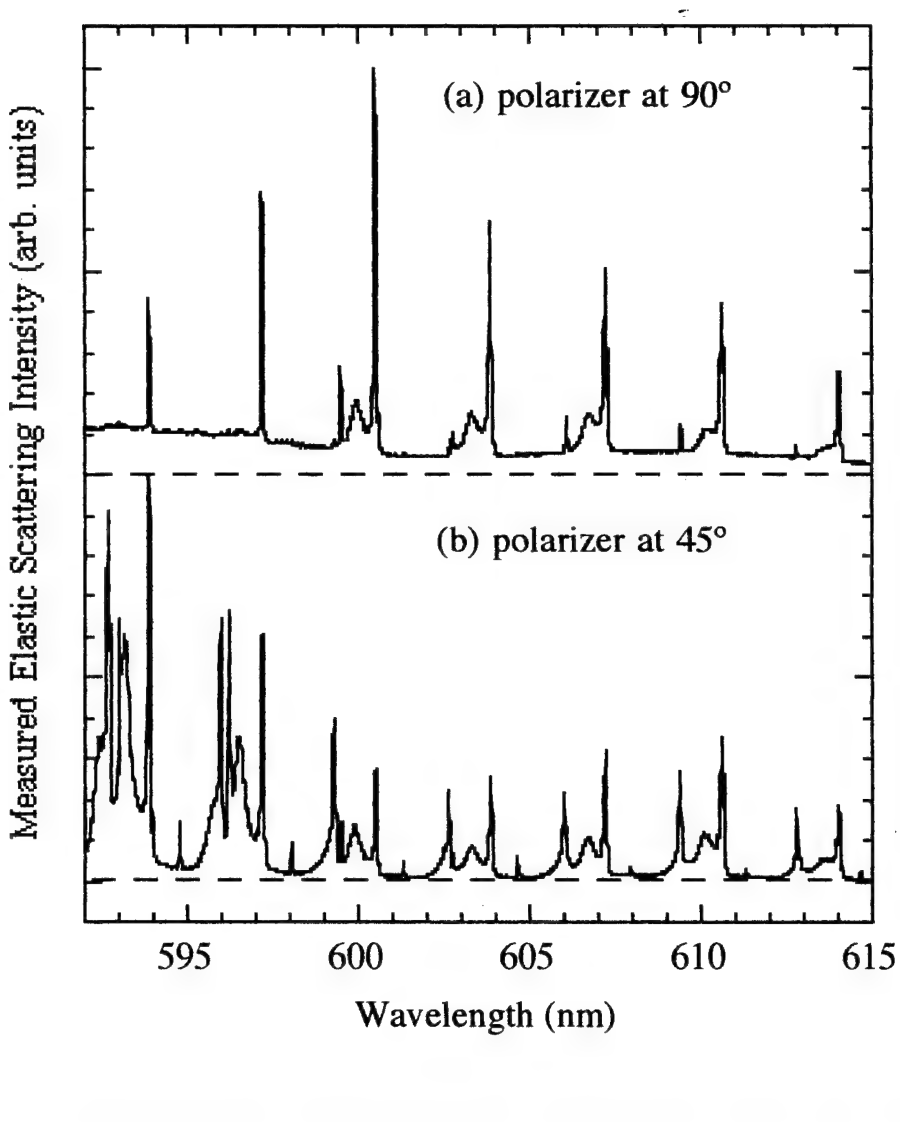

Fig. 7 Elastic scattering spectra for two orientations of the output polarizer (Fig.7 a selects only TE modes whereas Fig. 7 b selects both TE and TM modes).

index matched spectrum are replaced with significantly narrower resonances. In fact, the narowest of these resonances have widths only slightly larger than the dye laser resolution, $\lambda/\delta\lambda = 24{,}000$. These are clearly resolution limited. Deconvolution suggests Qs above 10^5, and Mie theory limits the value to 10^7. More important, there is essentially no backround in these spectra; the ray paths which normally lead to the majority of scattering for plane waves (Fig.3) are cut off.

Preliminary experiments showed that particles can be placed one behind another on the fiber coupler and be separately addressed. Although this was not possible for plane wave illumination, as demonstrated by our levitation experiments,[10] the elimination of the forward scattered field makes independent addressing viable.

Attempts to observe dips in the light transmitted through the fiber with our low resolution dye laser proved futile. Inorder to obtain higher resolution we utilized a distributed feedback semiconductor laser (DFB) with a linewidth of 10 MHz. Fig.8 shows the setup.[17]

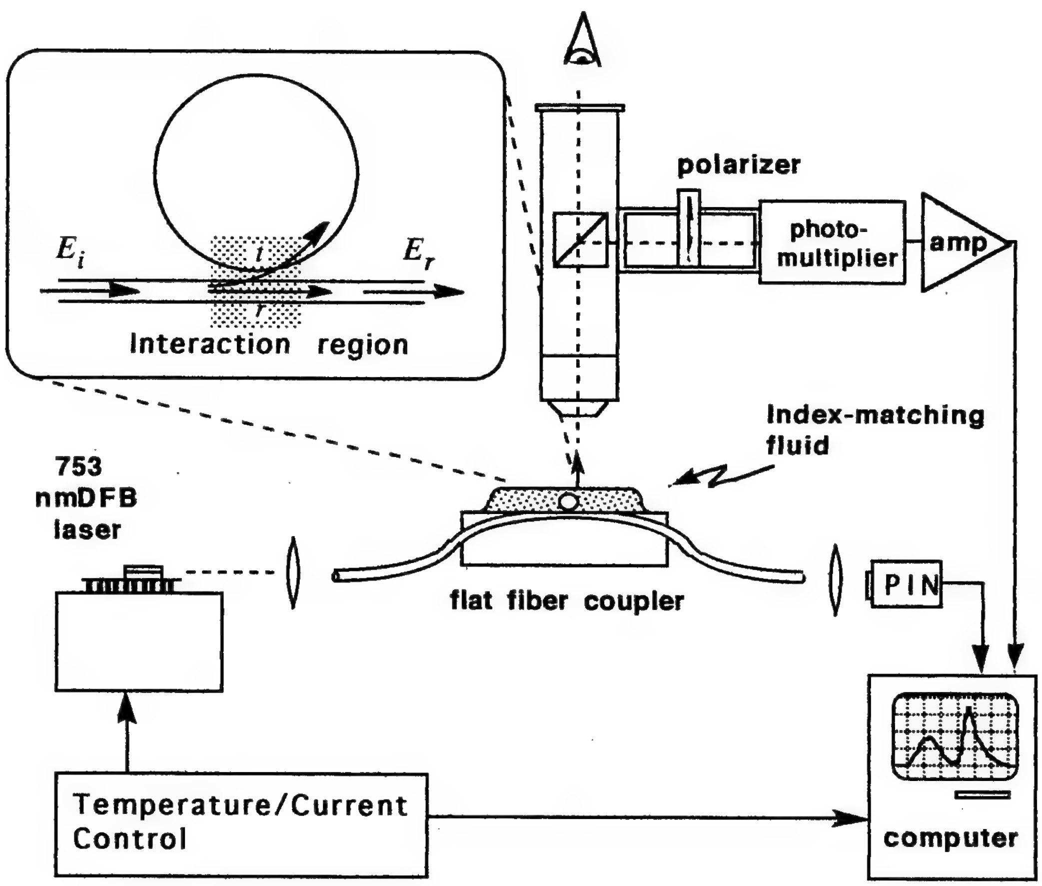

Fig. 8 High resolution setup for detetion of MDRs in both scattering and transmission.

The DFB laser was tuned by varying the drive current. Typical results using this device are shown in Fig. 9 for a large (a=500μm) index matched sphere composed of BK-7 glass. The dips are clearly present,[17] although their relative depths are currently unexplained.

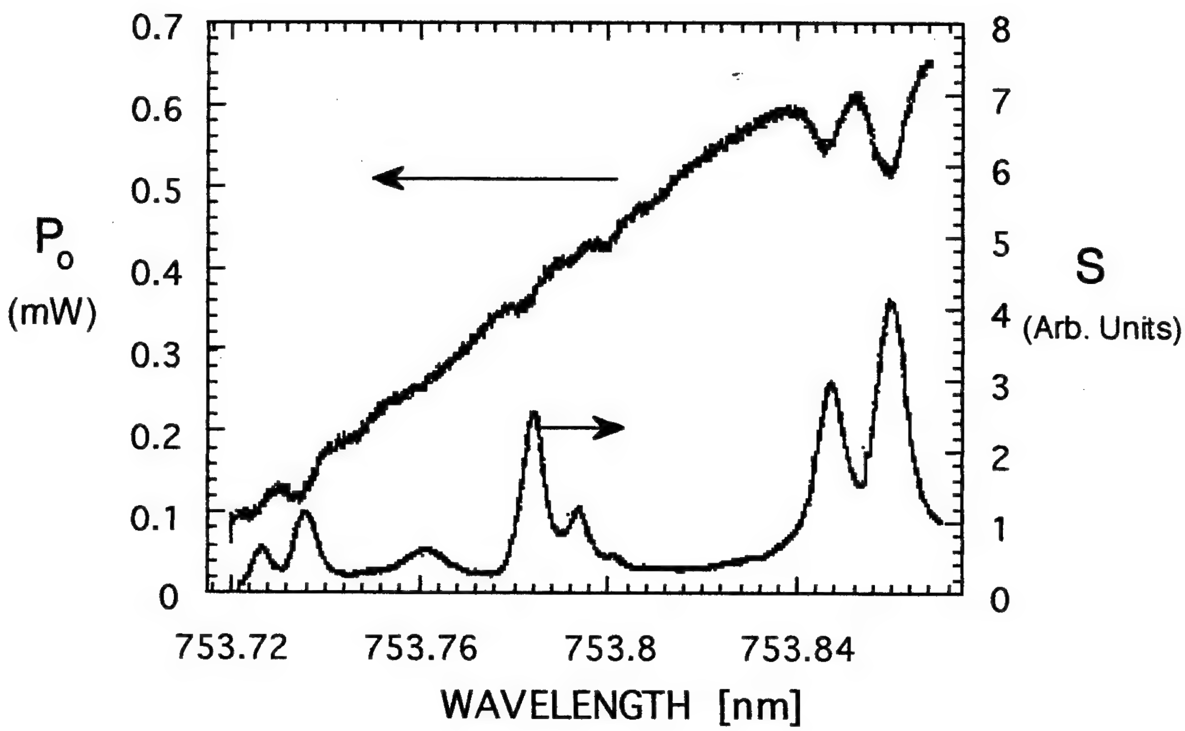

Fig. 9 TE spectrum of an index matched sphere taken by current tuning the DFB laser in Fig.8.

IV. SUMMARY and CONCLUSIONS

Microparticle Hole-burning Spectroscopy using plane waves is limited by multiple scattering initiated by rays which interact with spheres at low impact parameters, b<a; these rays do not excite spherical MDRs. The problem is overcome by coupling microspheres to the evanescent field associated with guided waves; evanescent fields couple principally to MDRs. This has been recently confirmed by numerical calculations which followed our original experiments.[18] In addition our own

preliminary experiments show that two particles can be placed one behind another on an optical fiber and be separately addressed. Although this was not possible for plane wave illumination, as demonstrated by our levitation experiments,[10] the elimination of the forward scattered field makes independent addressing viable.

Aside from Microparticle Hole-burning Spectroscopy our work opens the way to a host of new studies. It is possible, using evanescent coupling, to inject photons directly into high Q modes of active spheres (e.g. Nd-glass) where Cavity Quantum Electrodynamic (CQED) effects can lead to extremely low threshold lasing.[19] Even as passive elements, microspheres coupled to semiconductor lasers have the potential for stabilizing the output frequency and quenching the emission linewidth .

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VI. PUBLICATIONS, PRESENTATIONS and THESES

A. Publications

(i.) refereed

a. Published

1. S. Arnold, A. Ghaemi, and K.A. Fuller,
   Morphological Resonances Detected from a Cluster of two Spherical Particles,
   Opt. Lett. 19, 156-158(1994).

2. A. Serpenguzel, S. Arnold and G. Griffel,
   Excitation of Morphological Resonances from Individual Microparticles and Clusters in Contact with an Optical Fiber,
   Opt. Lett 20, 654-656 (1995).

3. G. Griffel, S. Arnold, D. Taskent, A.Serpengüzel, John Connolly, and Nancy Morris,
   Excitation of Morphological Resonances from Individual Microparticles and Clusters in Contact with an Optical Fiber,
   Optics and Photonics News, 6, 21 (1995).

4. G. Griffel, S. Arnold, A. Serpenguzel, S. Arnold and ,
   Excitation of Morphological Resonances from Individual Microparticles and Clusters in Contact with an Optical Fiber,
   Opt. Lett. 21, 695-697 (1996).

b. In Press

c. Submitted

1. A. Serpenguzel, S. Arnold and G. Griffel,
   Coupling of Guided Waves in Optical Fibers to Microsphere Resonances, submitted to JOSA B (July, 1996)

(ii.) other

a. Published

1. Microparticle Photonics: Fiber Optic Excitation of MDR's
   S. Arnold, A. Serpenguzel, and G. Griffel
   In *Guided Wave Optoelectronics*
   Ed. by T. Tamir, G. Griffel, and H.L. Bertoni
   (Plenum, New York, 1995)

2. Photonic Atoms: Enhanced Light Coupling
   A. Serpenguzel, S. Arnold and G. Griffel
   in *Microcavities and Photonic Bandgaps: Physics and Applications, Kluyver Netherlands (1996)*
   Ed. by J.G. Rarity (NATO Publications, Belgium, 1996)

B. Invited Lectures, Seminars, and other Presentations

a. Colloquia and Seminars Given at other Universities and Laboratories

1. City College of CUNY, December 14,1994
   "Photonic Atoms, Molecules, and Spectral Optical Memory"
   Physics Colloquim

b. Presentations at National and International Meetings

Invited

1. Guided Wave Optoelectronics (Invited)
   Microparticle Photonics: Fiber Optic Excitation of MDR's
   Polytechnic University, Brooklyn, New York, Oct. 28, 1994
   S. Arnold , A. Serpenguzel, G. Griffel

2. IEEE Frequency Control Conference (Invited)
   Quenching of Semiconductor Lasers Linewidth by Detuned Loading using Spherical Cavities Morphology Dependent Resonances
   San Francisco, CA, May 29-31, 1995
   G. Griffel, A. Serpengüzel, S. Arnold

3. Japan- U.S. Seminar(Invited)
   "Fluorescence Microscopy and Spectroscopy of Levitated Microparticles and Clusters: QED effects and more"
   Sept. 12, 1995, Hakone, Japan
   S. Arnold

Contributed

1. OE/LASE '95
   Excitation of Photonic Atoms (Dielectric Microspheres) on Optical Fibers: Application to Room Temperatire Persistent Spectral Hole-burning.
   San Jose, CA, Feb.4-10, 1995
   A. Serpenguzel, S. Arnold , G. Griffel

2. QELS'95 (International Conference Quantum Electronics)
"Spatially Selective Excitation of Dielectric Photonic Atom Resonances "
Baltimore, Md., May 22-26,1995
A. Serpengüzel, S. Arnold, and G. Griffel

3. LEOS'95
"Fiber Coupling of DFB Laser to Micro Spherical Cavities-A Novel Approach for Frequency Control and Linewidth Quenching Utilizing Morphology Dependent Resonances"
San Francisco, CA, October 31, 1995
G. Griffel, D. Taskent, S. Arnold, A. Serpenguzel, J. Connolly and N. A. Morris

4. 1996 ERDEC Conference on Obscuration and Aerosol Research
"Excitation of Morphology Dependent Resonances in Particles in Contact with a Thinly Clad Optical Fiber"
June 26-27, 1996, Aberdeen, Md.
S. Arnold, A. Serpenguzel and G. Griffel

5. 80th Annual Meeeting, Optical Society of America, October 20-24, 1996
"Photonic Atoms: Enhanced Light Coupling"
October 23, 1996, Rochester, NY
A. Serpenguzel, S. Arnold , G. Griffel

C. Dissertations and Theses

Student	Dept.	Date Begun	Completion Date
Ali Ghaemi	Physics		6/95